

PB-216 883

RADIOACTIVE CONTAMINATION AS A FACTOR IN
STREAM SANITATION STUDIES

O. W. Kochtitzky, Jr., et al

Public Health Service
Cincinnati, Ohio

31 August 1950

DISTRIBUTED BY:

NTIS

National Technical Information Service
U. S. DEPARTMENT OF COMMERCE
5285 Port Royal Road, Springfield Va. 22151

RADIOACTIVE CONTAMINATION AS A FACTOR IN STREAM SANITATION STUDIES

by

O.W. Kochtitzky, Jr. and Oliver R. Placak

In most production operations, there are industrial wastes for disposal. Disposal methods are influenced by the characteristics of the individual wastes. Some wastes have intrinsic by-product value and may be processed and sold at a profit. Others, such as liquid dye and sulfur wastes, have no value and may be objectionable to the physical senses of sight, smell and taste, or may be deleterious to aquatic life; while still others, such as sewage, may constitute a hazard to health. In industrial operations, these wastes may require expensive treatment, which must be considered as a part of the production cost. Neglect of such treatment, with liquid wastes, gives rise to stream pollution and public health problems.

Radioactive liquid wastes have no intrinsic value at the moment. They are not ordinarily perceptible nor objectionable to the senses. As normally encountered, radioactive wastes have no effect on aquatic plants or animals similar to the spectacular effects of exposure to toxic chemicals. True mutations or malformed fish have been produced by direct irradiation, but in the normal course of radioactive waste disposal, aquatic organisms do not appear to suffer obvious harm. Some organisms, such as algae, are very tolerant and appear to thrive in the presence of radioactive wastes. Such wastes do not transmit diseases

Reproduced by
NATIONAL TECHNICAL
INFORMATION SERVICE
U S Department of Commerce
Springfield VA 22151

U. S. PUBLIC INFORMATION SERVICE
ENVIRONMENTAL HEALTH CENTER

Reproduced from
best available copy.

as typhoid fever and dysentery are spread through the medium of sewage. Rather, any effects are more subtle but none the less important to health. It is true that very large doses could cause immediate injury or death but the concentrations to be expected in normal radioactive waste disposal would do neither. One would expect a long period of exposure to elapse before manifestation of any injury. Radioactive liquid wastes are a potential public health problem because continued or repeated exposure by immersion, ingestion or consumption of plants or animals which have concentrated radioactivity may result in accumulation within the body in sufficient quantity to induce harmful effects. The probability of these wastes becoming a stream pollution or health problem is enhanced by the fact that no treatment except natural decay or extreme dilution, without subsequent reconcentration, will reduce them to innocuous levels. All other treatment procedures result in a solid, gaseous or more concentrated liquid waste, containing equivalent radioactivity and a new disposal problem. The gain is only in volume and consequent facilitation of retention and storage.

To survey the role of radioactive wastes in stream pollution studies, one should be able to predict authoritatively the ultimate fate of these materials. At the present time, this is not as clearcut as the determination of more familiar criteria such as the deposition of sediment or the determination of oxygen depletion. At these levels, concentrations cannot be determined chemically and techniques and instru-

mentation not heretofore used in stream sanitation studies are required. For a complete appraisal of the situation even these marvelously sensitive instruments and techniques are still inadequate.

Radioactivity in water may be dissipated by decay. For some isotopes this is accomplished very quickly and for others is a slow process. For mixed isotopes, the nature and specific amount present must be known to predict reduction by decay accurately. Dispersion may result from dilution, absorption or adsorption. Thus, not only the wastes, but also the sediment, aquatic life both plant and animal, and immersed objects and structures may be involved. Where the radioactivity is bound in organic material the difficulties encountered in obtaining a true measurement are formidable. If all conceivable samples could be assayed, it is doubtful that radioactivity detected could be balanced against radioactivity added even if the latter values were known. One cannot, at the moment, predict accurately the ultimate fate of all radioactive materials if released to nature.

It should be obvious, that all statements made represent best judgment derived from experience to date. They may be modified as future developments contribute additional knowledge, new instrumentation and improved techniques.

Conventional Stream Survey Programs

Stream surveys are conducted for either investigative or regulative purposes. The method of approach and extent of the survey

- 4 -

are governed by the purpose. Surveys may be very specific as for example, the determination of the effect of a single waste or sewage plant effluent upon a limited stretch of river. The other extreme is a comprehensive appraisal of an entire river system. To accomplish the latter objective would require a study of the whole watershed, determination of all sources and quantities of objectionable materials and an evaluation of the effects of these materials with regard to domestic, recreational, agricultural and industrial use of the water.

Very broadly stated, a stream survey program would consist of the following items:

1. Data on wastes including source, volume, concentration, point of entry and nature of each pollutant.
2. Data on receiving stream including flow data, physical characteristics such as shape and condition of channel, temperature, stratification, impoundment, geological formations and short circuiting.
3. Data on downstream uses and for potential deleterious effects to agriculture, recreation, industry and domestic water supplies.
4. Data gathering, including judicious selection of sampling stations and also pertinent chemical, biochemical, biological, and bacteriological analyses.

5. Data reporting and recommendations. This will include assembly and compilation of data for the records and informational value, interpretation for suggesting remedial measures and if necessary use of the data to enforce compliance with existing legislation.

Obviously, if we are faced with wastes containing radioactive materials, these must also be fitted into this pattern. Reconsideration of the concise statement of a conventional stream survey program will indicate no intrinsic reason against the incorporation of radiological studies in such a program. Some practical difficulties are apparent. Any such difficulties are due primarily to the newness of the field. They are not insurmountable and will decrease with the development and standardization of techniques, availability of more suitable measuring instruments and increases in knowledge and trained personnel. Some of these factors stated briefly, are:

1. For obvious reasons, data at the source may be unavailable.
2. Knowledge concerning physiological and biological factors is still incomplete and therefore maximum permissible concentrations are not absolute values.
3. Techniques and analytical methods have not been completely developed or standardized.
4. The ultimate fate in nature of these materials needs elaboration.

5. Knowledge is required regarding the role radioactive materials may play in biological food chains and subsequent reconcentration.
6. At the concentrations normally encountered, chemical analysis may be impractical and interpretation of results less certain.
7. Many of the elements involved were previously of little concern in nature and consequently no back log of information is available.
8. The personnel normally engaged in stream survey work must be indoctrinated sufficiently to understand the terminology and implications of this potential new factor.

Sources of Radioactive Materials

If radioactive materials were encountered in a stream, they would most surely originate from one or more of the following sources:

1. Naturally radioactive materials
2. Operating nuclear reactors
3. Hospitals or research institutions
4. Industrial use
5. Atomic explosion

Natural background radioactivity is a base line to which all survey studies will have to be related and over which we have no control. The accurate determination of radioactivity at these low

levels requires special techniques and skilled personnel. Operating nuclear reactors are the greatest sources of radioactivity in quantity and variety and the ones in which the least information will be available at the source, at least until we live in a more settled world. Hospital, research institution and industrial users should fit into a normal pattern and information on wastes at the source will be available. In general, they will use specific or a limited number of isotopes which will simplify interpretation of results. G.H. Guest⁽¹⁾ has published an article describing present industrial use of radioisotopes. Thirty-seven potential uses are given and perusal of these will suggest those fields which may contribute radioactive wastes in the future. Atomic explosions are atypical and not to be considered in normal operations.

Probable Fate of Radioactive Contaminants in Nature

With present knowledge, it is impossible to state exactly and quantitatively the ultimate destination of all radioactive materials that might enter a body of water. One can, however, visualize and describe the mechanism in broad terms.

For the purpose of this delineation, we may assume that appreciable quantities of radioactive materials are present, a situation that permits the widest possible latitude, in description. Even if the radioactive materials were present in less than maximum

permissible concentrations they would follow the same pattern but of course might be difficultly detectable and entirely different in their public health and other implications.

Since radioactivity can neither be neutralized nor destroyed, water-borne radioactive materials will be removed and concentrated in some manner or remain in the liquid phase.

The sole positive and permanent form of removal is through the mechanism of natural decay. This characteristic is inherent in a given isotope and varies from seconds to millions of years. Physical removal from water may be accomplished by mechanical or chemical precipitation, absorption by biological organisms, adsorption on surfaces such as clay particles, deposition on structures such as bridge piers and boats, assimilation into plants or any combination of the above mechanisms. Regardless of the manner of removal, the end results are new foci of radioactive materials which need investigation and evaluation. If, for example the mode of removal is incorporation in organized biological material, such as algae or fish, the resulting concentration in the organism may be much higher than that in the environment in which the organism exists.

High concentrations of activity at a point remote from the source of the wastes can occur by other means than those outlined above. Channelization, thermal stratification, geological faults, and transportation of deposited material may prevent adequate dispersion and dilution.

The radioactive isotopes remaining in the liquid phase would persist unless removed by some intervening process, such as a water treatment plant, until they become undetectable through dilution or decay. Still assuming appreciable amounts of radioactive contaminants present, that portion in the liquid phase and to some extent those adsorbed, deposited or concentrated (depending on the point of deposition or concentration) would primarily create public health problems. These effects would result from external exposure, as in swimming, or internal exposure through ingestion of water or food and possible fixation in the body. The magnitude of the effect would vary with the isotope and would depend on concentration, half-life (both biological and radioactive), energy and type of radiation, chemical properties and mode of metabolism. In general, one would be most concerned with isotopes of intermediate half-life. With long half-lives the specific ionization would be low and with very short half-lives the integrated dose would be low.

If this water became a source of supply for domestic use, considerable removal could be expected during the treatment process. The efficiency of such removal varies with the isotope and would probably range from 95-100% with P^{32} and the rare earths to less than 10% in the case of I^{131} and Sr^{89} . In the first case, that of efficient removal, it is obvious that the process is also an efficient concentrator of radioactivity and the end result is contaminated sludges. In the latter case, these materials would pass through the plant and enter the

distribution system, where they would have to be considered as ingestion hazards. The final step in the cycle would be entry into the sewers, processing and concentration in the municipal sewage plant and sewage sludges and re-disposal of the effluent in the river. Improper or accidental discharge of radioactive wastes into sewers, by non-commission users, would also contribute to the contamination of sewage processes.

If, contaminated water were to be used as industrial or process water, the possibility of contamination of equipment and products would have to be considered. A striking example would be the manufacture of paper to be used later for packaging photographic films.

Supplementary Samples and Analyses Necessitated
by Radioactive Contaminants

In the presence of radioactive contaminants, biological samples and physical samples, which ordinarily would not be collected, are more significant than during normal stream studies. Consequently, more attention should be devoted to the collection, analysis and significance of these samples.

Microscopic organisms, plankton and algae may concentrate radioactivity by a factor of several thousand over their water environment. When utilized as fish food, they are converted to a form suitable

for human consumption. If not used in this manner, they may die and become part of the bottom sediment or possibly be removed by a water plant contributing to the radioactivity in the sludge or filter wash water. Macroscopic organisms (fish, worms, snails, aquatic animals) can be examined for radiation damage or assayed for absorbed radioactivity. Aquatic vegetation may also be examined for radioactivity.

Suspended solids or bottom sediments in general, adsorb radioactivity and may have associated with them concentrations of radioactivity in excess of that present in the liquid phase. Such radioactive solids would settle out, be removed in water plants, possibly build up in recreation areas or enter into the formation of deltas.

In the presence of significant amounts of radioactivity, one might suspect adsorption on marine structures, bridge piers and boats or these structures could have adherent radioactive muds or algae.

It is possible for the concentration of radioactive contamination to occur in the flocs and sludges of water and waste treatment plants and in the slimes and sediments of pipes, conduits and other hydraulic structures. With mixed fission products, one would suspect that 70-80% of the radioactivity entering a conventional water plant would be removed and concentrated within the plant. One should

bear in mind, though, that specific isotopes might leave the plant essentially unchanged in concentration.

Since our knowledge of the dispersion and behavior of radioactive contaminants in nature is still deficient in many respects, one might consider the usefulness of all the types of samples suggested.

Instrumentation

Since radioactivity cannot be detected visually or chemically in a simple manner, but requires electrical or photo sensitive devices, the procedures and instruments for investigating and measuring contamination are highly specialized and entirely new to the field of stream sanitation studies. Popular literature has publicized the availability of Geiger-Muller detectors at nominal prices. Such instruments are hardly adequate for measuring levels of radioactivity in the range of tolerance values. Instruments for such measurements are generally complex electronic circuits and detectors and are both expensive and require judgment in operation. Improvements and adaptation of commercially available instruments are frequently necessary for specific uses.

At Oak Ridge National Laboratory, a number of highly specialized probes or detectors, for field use, have been or are being developed. One such instrument is the so-called "flounder" for measurement of radioactivity in the water of streams or lakes.

This probe consists of twelve G-M tubes enclosed in a flat water-proof plastic case. It has been calibrated with known sources and can actuate a counting rate meter to indicate intensity (milli-roentgens per hour) but not concentration. To quantify the observation for microcuries per cubic centimeter would require knowledge of the total effective water volume, energy of emission, distance of all increment volumes from the detector and the quantity and absorptive capacity of intervening suspended matter. To incorporate all of these corrections into a single instrument is virtually impossible when mixtures of unknown radioelements are involved. The instrument, however, is very versatile. Using different wall thicknesses a distinction can be made between gamma and beta radiation. By resting the "flounder" on the bottom and correcting for the activity of the water, the intensity at the mud surface may be estimated. When towed through the water, areas of concentration, stratification and channelization may be detected.

In addition to the "flounder", a mud probe has been designed which can be pushed into the bottom sediment to measure intensity at different depths. This probe consists of a specially designed G-M tube, attached to an aluminum pipe with linear markings for the measurement of depth and mud penetration. This instrument must be sturdy enough to withstand rough field use and consequently the walls of the stainless steel tube are of such thickness as to pass only gamma radiation.

Stainless steel tubes similar to the mud probe but without pipe extensions have been devised for field use as fish probes. These are similar to, in some respects but more rugged than, the surgical probes which are commercially available. A thick-wall tube for gamma and a thin-wall tube for beta are attached to a portable battery operated scaler and used alternately by a switch arrangement. Fish caught in nets and held in a tub of water may be monitored by insertion of the tube into their throats and subsequently returned to the water without harm. The probe can be calibrated only by the sacrifice and assay of a number of fish after probing. While insufficient work has been done, we suspect the calibration will vary with size and species of fish because of varying proportion of flesh, bone, and scales and the capacity of different tissues to concentrate radioactivity. Furthermore, the calibration may vary with the specific isotopes present in the wastes and the chemical composition of the fish, the available food supply, and the water.

Well probes have been constructed for use in test wells to determine percolation of ground water. This instrument consists of a G-M detector and pre-amplification circuit enclosed in a waterproof case attached to the end of a long electric cable. It may be lowered from a reel at a constant rate into the well and through the counting rate meter and strip recorder produce a continuous record of intensity of radiation from top to bottom. The U.S. Geological Survey has similar instruments and there are commercial enterprises in the business of

gamma well logging. The commercial operations, however, are for quite a different purpose. By determining the gamma radiation from the rocks penetrated in a well a geological log can be produced which will give indication of the presence of oil bearing strata.

Continuous monitoring presents intriguing possibilities. To date, however, no completely satisfactory immersion detector for continuous monitoring under stream conditions has been devised. The difficulty is one of fouling. Algae and turbidity accumulating on the detector invalidate the records. Efforts are being made to develop an effective anti-fouling coating to obviate this difficulty. Detectors have been suspended over the flowing water with more success. However, fluctuation in water levels creates variable distances from the water surface to the detector and results in inconsistent intensity detection. Furthermore, detectable radiation from nearby structures varies with the amount of surface exposed and this factor is governed by variations in water level. Efforts are being made to develop means for suspending detectors in such a manner that they will remain at a fixed distance from the water surface regardless of change in water elevation and at the same time provide shielding from nearby contaminated surfaces (banks, structures, etc.).

Some of these problems are non-existent in the case of constant monitoring of the effluent water from a purification plant. Here the suspended matter and algae can be removed so that they will not foul the detector and the distance from water to detector can be kept

constant. The level of activity, however, may be such that large quantities of clarified water must be passed in thin films under or around the detectors in order to get a significant measurement. Thought is being given to concentration of activity by flocculation or evaporation in order to produce more readily measureable concentrations.

Since constant monitoring is not quantitative when unknown radioelements are present and is wrought with complicating interferences, continuous sampling for subsequent analysis may be employed. Many sampling devices are available and the sampler should be selected for the specific needs involved. For constant and uniform flows a simple continuous or intermittent sampler may be employed but where wide variations in flow are experienced arrangements for proportional sampling should be employed. This is relatively simple for pipeline or weir flows but surface streams with great variation in elevation as well as flow may involve elaborate arrangements if these devices are used.

Gross Analysis

Most analyses in a stream survey program will consist of gross counts of whatever mixture of isotopes may happen to be present. For gross analysis a sample of small thickness (of the order of milligrams per square centimeter) to minimize self absorption is deposited on a dish or disk, exposed to a counter and the total count for a definite

time interval determined. If the thickness used is such that a portion of the radioactive emanations are absorbed in the sample itself, a correction must be applied. Counting geometry (ratio of counts observed to true count) is sensitive to the energy of the emanation counted. A gross count from a mixture of isotopes of different energies will require strict interpretation to quantify to such units as micro-curies per cubic centimeter. Since the level of radioactivity in these samples will ordinarily be low, and no counter will count all disintegrating atoms, the activity in many samples must be concentrated by evaporation, precipitation or other methods. It is obvious that the level of activity will determine the sample size. In our laboratories, we have concentrated as much as 50 gallons of well water and 300 gallons of rain water to obtain significant counts. Of course, at such levels the concentration is very much below tolerance values.

Counting tubes of different wall thickness or used in conjunction with separate absorbers will discriminate between different types of radiation. Thus it is possible to screen out betas and count only gamma activity. Such devices as proportional counters or open window or extremely thin-walled detectors would be needed for the evaluation of alpha radiation.

A simple count of disintegrating atoms or a known proportion thereof will give a measure of total radioactivity. If only one isotope is present, or a very simple mixture, identification may be accomplished

by energy and decay time measurements. Energy values are obtained by evaluating a series of measurements made with increasing thicknesses of absorber interposed between the sample and the detector. Decay time is determined by repeated counts at successive time intervals to either observe or calculate the time required to reduce the counting rate by one half.

Radiochemical Analysis

In the presence of a number of radioisotopes such as mixed fission products, identification is more difficult. Where this is necessary, chemical separation of the various elements, known or suspected to be present, will be required. Such procedures are time consuming and tedious and require techniques unfamiliar to those normally engaged in stream survey work. At the levels to be expected, complete analysis may be impossible. Some compromise, such as identification of the main constituents, possibly 70-80% of the total, would have to be accepted. For a radiochemical analysis of mixed fission products in our laboratory, we would prefer the activity level of the samples to be at least 500 counts per minute per milliliter or capable of concentration to this level for analysis. Analysis at lower levels would be attempted, if the occasion warranted, but with less confidence in the validity of the determination. Where radiochemical assay is required a competent radiochemist should be employed or as a minimum present personnel should be given thorough training in radiochemical techniques and procedures.

Autoradiography

In addition to assay and analysis, solid specimens can be examined by autoradiography. In this technique specially produced films are exposed to the specimen for a period of time, maybe days. Many things can be learned from autograms but their production and interpretation is a highly specialized field requiring long training and experience.

Appraisal of Results

No stream study program is worthwhile unless one compiles, analyzes and reports the results with recommendations for improvement. In general in a complex environmental health program, certain fundamental elements will always have to be considered. Briefly stated, they are:

1. Recognition of danger to health, determination of causes and establishment or adoption of maximum permissible limits of exposure.
2. Determination of where, how and to what extent the harmful agent may occur.
3. Devise methods to control the hazard at its source or eliminate or minimize its effects in the environment.

4. Measure and appraise the effectiveness of control measures after they have been established, and re-examine evidences of injury to health to be sure the expected benefits to health are realized and are dependable.

For radioactive contaminants, as in the case of other contaminants, the data should be interpreted to fulfill these requirements, insofar as possible. The data obtained in a well conducted survey will determine the source, concentration and location of the contaminant. The results must be interpreted in terms of human or other hazard.

There are no universally accepted maximum permissible concentrations. When these become available they should be adopted as criteria. Two subcommittees of the National Committee on Radiation Protection are endeavoring to establish these values. They are the subcommittees on "Permissible Dose from External Sources" and "Permissible Internal Dose". In the interim established working tolerances should be used. For mixed fission products, the value 10^{-7} uc/cc⁽²⁾ should be used. Where the radiation is known to be from a specific isotope, the value for that isotope should be used. This may vary, by several orders of magnitude from the general value. Some currently proposed maximum permissible values for use in internal exposure are given by Morgan⁽³⁾. The values in use for external radiation are given in Handbook 42⁽²⁾, "Safe Handling of Radioactive Isotopes", and this

is recommended reading for its brief discussion of the elements that determine radiotoxicity and the classification of radioisotopes according to relative radiotoxicity.

In addition to the discussion contained in Handbook 42 some other factors should be considered.

1. Alpha radiation will seldom constitute an external radiation hazard.
2. Obviously, effects of radioactive materials retained in the bottom sediment will be of a different order of magnitude and represent a different kind of hazard from similar activity contained in the liquid phase.
3. Radiation exposure is cumulative, and while water alone may not be above tolerance values, a summation of radiation from all sources may be above such values. At the concentrations to be expected in stream contamination, total exposure over a long period of time is probably more significant than momentary exposure.
4. All biological organisms do not react in the same way and there is a possibility of radiosensitive individuals.
5. Where isotopic dilution is a factor, the uptake of radioactive materials will be limited.

6. The concentration in water will have to be interpreted in the light of potential biological or physical reconcentration.
7. Where standards for human tolerance are met all other uses will probably be taken care of, with the possible exception of contaminated materials that would increase background in experimental equipment.

The regulation of the disposal of radioactive wastes has been a function of the Atomic Energy Commission either in its own installations or through licensing of non-commission users of isotopes. A popular document published by the Commission regarding radioactive wastes (4), "Handling Radioactive Wastes in the Atomic Energy Program", is recommended reading. Few industries have been as conscientious in attempting to solve an industrial waste problem. Inevitably, however, as with other wastes it may be presumed that appraisal of injury and regulation will be entrusted to public health authorities. As criteria for methods of disposal at the source, the data available in commission installations will be invaluable. Other sources of information will soon be available and should be used, such as the findings of the subcommittee on "Waste Disposal and Decontamination of the National Committee on Radiation Protection".

References

1. Guest, G.H. - Industrial Uses of Radioisotopes - CRIB-399.
National Research Council of Canada.
2. Handbook 42 - Safe Handling of Radioactive Isotopes. U.S.
Department of Commerce, National Bureau of Standards.
3. Placak, O.R. and Morton, R.J. - Research on the Disposal of
Radioactive Wastes. J.A.W.W.A. 42, No. 2, pp 135-142 (February
1950).
4. Handling Radioactive Wastes in the Atomic Energy Program - U.S.
Atomic Energy Commission.

OWK/ORP:r
August 31, 1950